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(54) Title: HOMOGENEOUS THERMOSET COPOLYMERS

(57) Abstract

Novel compositions of matter comprise homogeneous thermoset copolymers resulting from the reaction between a poly(vinyl benzyl ether) of a polyphenol and a cyanate compound which may be a dicyanate ester of a polyether of a polyphenol or a polyaromatic cyanate compound which may be used as components in laminates which are employed in electronic circuit boards. The copolymers will impart desirable characteristics such as high glass transition temperatures and low dielectric constants to the finished product.

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HOMOGENEOUS THERMOSET COPOLYMERS

BACKGROUND OF THE INVENTION

With the advent of sophisticated equipment in the electrical and electronic fields, it has become necessary that the components of the various pieces of equipment conform to high standards which are set forth in the specifications for these components. For example, circuit boards which are used in relatively complicated pieces of equipment such as main frame computers, must be of a relatively high standard of quality in order to function in an efficient manner for a long period of time without deteriorating or breaking down, and thus causing an interruption in the function of the machine. This high quality of material is opposed to pieces of equipment requiring a lower standard of quality such as those used in personal computers, high quality television equipment, radios, etc.

Circuit boards upon which a circuit is etched or implanted usually comprise a laminate which is composed of a synthetic polymeric substance which possesses desirable characteristics such as thermal stability, low coefficient of thermal expansion, dimensional stability, low dielectric constant, solvent resistance, low moisture absorption, etc. and a suitable reinforcement matrix, such as glass, quartz, graphite, Kevlar, etc.

As will hereinafter be shown, it has now been discovered that a homogeneous thermoset copolymer of a poly(vinyl benzyl ether) of a polyphenol and a cyanate compound which may be a dicyanate ester of a polyether of a polyphenol or polyaromatic cyanate compound may be used in the preparation of laminates which themselves will form a component of a circuit board and will possess the desirable characteristics hereinbefore set forth.

BRIEF SUMMARY OF THE INVENTION

This invention relates to homogeneous thermoset copolymers. More specifically, the invention is concerned with novel high temperature homogeneous thermoset copolymers and to a method for the preparation thereof. As was previously mentioned, the homogeneous thermoset copolymers of the present invention, which constitute novel compositions of matter, may be used to coat and/or impregnate a substrate which is thereafter cured and utilized in circuit board laminates and dielectric coatings, the use thereof being attributable to the desirable characteristics which are possessed by these

polymeric compositions of matter. The particular characteristics of the polymer dielectric and reinforcing components which go to make up the circuit boards contribute to the efficiency and stability of the final electronic equipment in which the circuit boards are used. For example, a lowering of the dielectric constant in the polymer matrix reduces the signal delay time or "crosstalk" and line capacitance. This results in faster PWB circuitry and, in addition, provides the potential to increase the number of functions per board. The polymeric matrix of the present invention possesses a lower dielectric constant than that which is possessed by thermosetting polyimide or epoxy matrices which are used as the standards by the industry for electrical laminates.

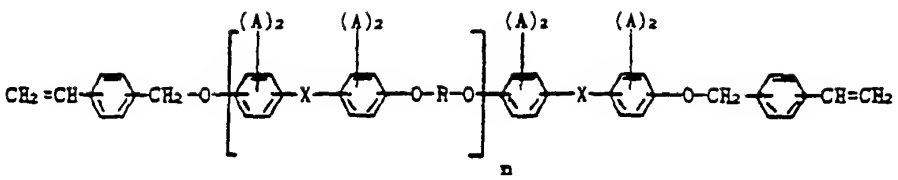
Another desirable characteristic of a polymer matrix for use in circuit boards is that the coefficient of thermal expansion should be relatively low in order to avoid a mismatch of thermal expansions with the electronic components and the fiberglass reinforcement with which the polymeric matrix is composited. It has been found that the coefficient of expansion of the novel homogeneous thermoset copolymers of the present invention is comparable to a polyimide matrix. Furthermore, the thermal stability of the polymer matrix must be relatively high in nature inasmuch as the matrix must possess the ability to withstand soldering temperatures without melting or degrading. A desirable characteristic of the homogeneous thermoset copolymer of the present invention is that the thermal stability of the polymer is comparable to a polyimide matrix.

In addition, by varying the ratio of the poly(vinyl benzyl ether) of a polyphenol to the cyanate compound, it is possible to provide a wide range of properties which will meet various and specific circuit board requirements. Furthermore, it is possible by preparing the copolymers in a manner also hereinafter set forth in greater detail, to provide a copolymer which will meet the requirement for chip encapsulation and potting materials.

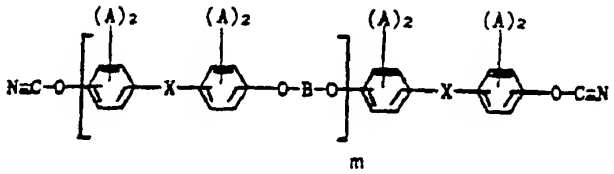
It is therefore an object of this invention to provide novel homogeneous thermoset copolymers.

Another object of this invention is to provide a method for preparing homogeneous thermoset copolymers which are useful as a component in circuit board laminates.

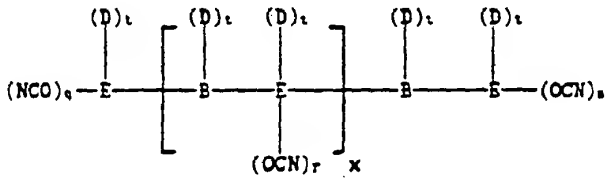
In one aspect, an embodiment of this invention resides in a homogeneous thermoset copolymer of a poly(vinyl benzyl ether) of a polyphenol having the structure:



in which X is selected from the group consisting of CH_3-$\underset{|}{C}$-CH_3, SO_2, O, S, and CH_2 radicals, R is selected from the group consisting of $-CH_2-C_6H_4-CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2-CH=CH-CH_2-$ and $-CH_2-C\equiv C-CH_2-$ radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and n has an average value in the range of from about 0 to about 20 and a cyanate compound selected from the group consisting of a dicyanate ester of a polyether of a polyphenol having the general formula:

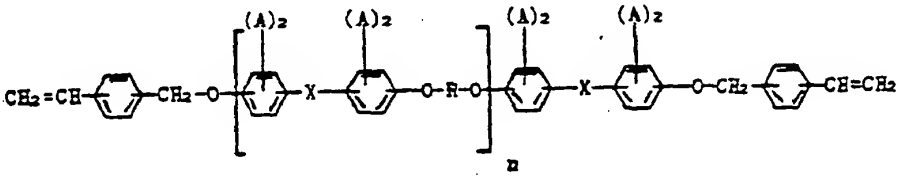


in which X is selected from the group consisting of CH_3-$\underset{|}{C}$-CH_3, SO_2, O, S, and CH_2 radicals, B is selected from the group consisting of cyclic and polycyclic aliphatic radicals containing from 5 to about 22 carbon atoms, $-CH_2-C_6H_4-CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2-CH=CH-CH_2-$ and $-CH_2-C\equiv C-CH_2-$ radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and m has an average value in the range of from about 0 to about 20 and a polyaromatic cyanate compound having the general formula:

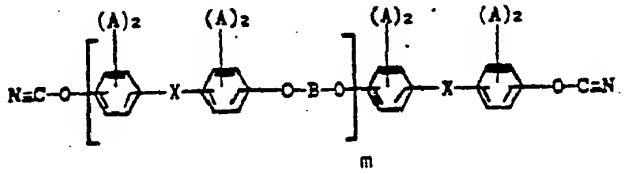


in which B is a polycyclic aliphatic radical containing from about 7 to about 20 carbon atoms, D is any nonactive hydrogen-containing substituent, E is an aromatic radical, q, r and s are independently in each occurrence the integers 0, 1, 2, or 3, with the proviso that the sum of q, r and s is greater than or equal to 2, t is independently in each occurrence an integer of between about 0 and 4, and x is a number between about 0 and 5.

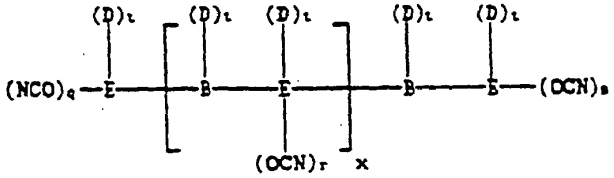
Another embodiment of this invention is found in a process for the production of a homogeneous thermoset copolymer which comprises reacting a poly(vinyl benzyl ether) having the structure:



in which X is selected from the group consisting of $CH_3\text{-}\underset{\underset{|}{}}{\overset{\overset{|}{}}{C}}\text{-}CH_3$, SO_2, O, S, and CH_2 radicals, R is selected from the group consisting of $-CH_2-C_6H_4-CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2-CH=CH-CH_2-$ and $-CH_2-C\equiv C-CH_2-$ radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and n has an average value in the range of from about 0 to about 20 with a cyanate compound selected from the group consisting of a dicyanate ester of a polyether of a polyphenol having the structure:



in which X is selected from the group consisting of $CH_3\text{-}\underset{\underset{|}{}}{\overset{\overset{|}{}}{C}}\text{-}CH_3$, SO_2, O, S, and CH_2 radicals, B is selected from the group consisting of cyclic and polycyclic aliphatic radicals containing from 5 to about 22 carbon atoms, $-CH_2-C_6H_4-CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2-CH=CH-CH_2-$ and $-CH_2-C\equiv C-CH_2-$ radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and m has an average value in the range of from about 0 to about 20 and a polyaromatic cyanate compound having the general formula:



in which B is a polycyclic aliphatic radical containing from about 7 to about 20 carbon atoms, D is any nonactive hydrogen-containing substituent, E is an

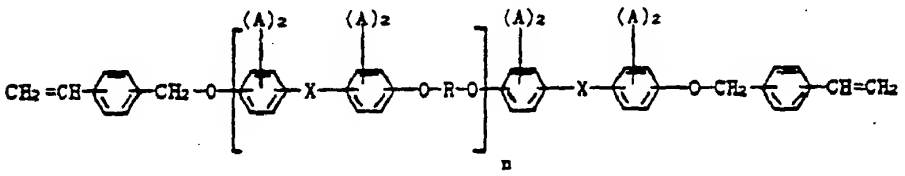
aromatic radical, q, r and s are independently in each occurrence the integers 0, 1, 2, or 3, with the proviso that the sum of q, r and s is greater than or equal to 2, t is independently in each occurrence an integer of between about 0 and 4, and x is a number between about 0 and 5 at reaction conditions, and recovering the resultant homogeneous thermoset copolymer.

Other objects and embodiments will be found in the further detailed description of the invention.

DETAILED DESCRIPTION OF THE INVENTION

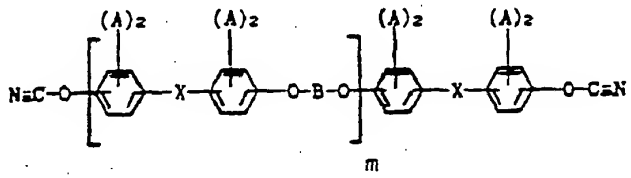
As hereinbefore set forth, the present invention is concerned with novel homogeneous thermoset copolymers and to a method for the preparation of these copolymers. The homogeneous thermoset copolymers will comprise a mixture of a poly(vinyl benzyl ether) of a polyphenol and a cyanate compound which may be a dicyanate ester of a polyether of a polyphenol or a polyaromatic cyanate compound, examples of these compounds being hereinafter set forth in greater detail. The copolymerization of these compounds will result in a copolymer which, because of its particular structure, will be useful in electronic circuitry, said composite possessing a relatively low dielectric constant and a high glass transition temperature, these characteristics being of particular advantage for use in circuit boards.

The poly(vinyl benzyl ether) of a polyphenol which forms one component of the copolymer of the present invention will possess the generic structure:

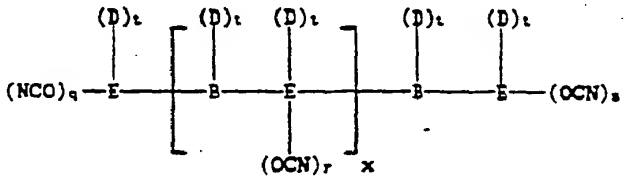


in which X is selected from the group consisting of $CH_3\text{-}\underset{\underset{CH_3}{|}}{C}\text{-}CH_3$, SO_2, O, S, and CH_2 radicals, R is selected from the group consisting of $-CH_2\text{-}C_6H_4\text{-}CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2\text{-}CH=CH\text{-}CH_2-$ and $-CH_2\text{-}C\equiv C\text{-}CH_2-$ radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and n has an average value in the range of from about 0 to about 20.

The second component of the copolymer of the present invention will comprise a cyanate compound comprising dicyanate ester of a polyether of a polyphenol which possesses the generic structure:



in which X is selected from the group consisting of CH_3-$\overset{|}{\underset{|}{C}}$-$CH_3$, SO_2, O, S, and CH_2 radicals, B is selected from the group consisting of cyclic and polycyclic aliphatic radicals containing from 5 to about 22 carbon atoms, -CH_2-C_6H_4-CH_2-, $(CH_2)_b$ in which b ranges from 1 to about 6, -CH_2-CH=CH-CH_2- and -CH_2-C≡C-CH_2- radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and m has an average value in the range of from about 0 to about 20 or a polyaromatic cyanate compound having the general formula:



in which B is a polycyclic aliphatic radical containing from about 7 to about 20 carbon atoms, D is any nonactive hydrogen-containing substituent, E is an aromatic radical, q, r and s are independently in each occurrence the integers 0, 1, 2, or 3, with the proviso that the sum of q, r and s is greater than or equal to 2, t is independently in each occurrence an integer of between about 0 and 4, and x is a number between about 0 and 5. In the above generic structure which refers to the dicyanate ester of a polyether of a polyphenol, the cyclic or polycyclic aliphatic radical referred to as B will be comparable in nature to those radicals set forth in U.S. Patent 4,528,366. Likewise, the definitions of the B, D and E radicals in the general formula for the polyaromatic cyanate component of the homogeneous thermoset copolymer are also comparable in nature to those definitions likewise set forth in U.S. Patent 4,528,366.

The novel homogeneous thermoset copolymers of the present invention may be prepared in any suitable manner of operation which is known in the art. For example, one method of preparing the copolymer is to blend the

two components hereinbefore described to form a homogeneous melt, said blending being effected at a temperature in the range of from about 100°C to about 200°C. The homogeneous melt may then be poured into a mold or utilized in other various ways such as encapsulating a microelectronic, integrated circuit chip, and cured at an elevated temperature in the range of from about 150°C to about 200°C and at a pressure from atmospheric to about 50 atmospheres for a period of time which may range from about 0.1 to about 1 hour, and thereafter may be post-cured at a temperature ranging from about 200° to about 260°C for a period of time which may range from about 1 to about 12 hours in duration.

Alternatively, another method of preparing the desired copolymer of the present invention is to dissolve the poly(vinyl benzyl ether) of a polyphenol and the dicyanate ester of a polyether of a polyphenol or the polyaromatic cyanate compound in an appropriate solvent such as dimethyl formamide, N-methylpyrrolidinone, dimethyl acetamide, acetone, benzene, toluene, etc., in amounts so that the resulting solution will contain from about 10 to about 90% by weight of the components of the copolymer. The resulting solution may then be coated and/or impregnated on an appropriate substrate such as various resins, glass cloth, etc., and treated at an elevated temperature of from about 150° to about 180°C for a relatively short period of time, which may range from about 1 to about 10 minutes, to obtain a prepreg. The resulting prepreg may then be stacked by pressing a predetermined number of sheets of the prepreg and pressing the stack in a heated press to form a desired laminate. The pressing of the prepreg may be effected for a period of time ranging from about 0.5 to about 4 hours in duration at an elevated temperature ranging from about 150 to about 190°C, at a pressure in the range of from about 200 to about 1,000 pounds per square inch gauge (1378 to 6894 kPa gauge). Following the pressing, the laminate is then subjected to a post-cure which is effected at a temperature in the range of from about 200° to about 260°C for a period of time which may range from about 1 to about 8 hours in duration.

It is also contemplated within the scope of this invention that the homogeneous thermoset copolymers may be prepared in a continuous manner of operation. When this type of operation is employed, the predetermined amounts of the poly(vinyl benzyl ether) of a polyphenol and the dicyanate ester of a polyether of a polyphenol or polyaromatic cyanate compound which have been dissolved in an appropriate solvent of the type hereinbefore set forth in greater detail, are continuously charged to a zone which is maintained at the proper operating conditions of temperature and pressure. Those skilled in the art will

recognize that a continuous reactant charge is necessary, with amounts depending upon the individual components, to provide a high yield of product which contains the desired percentage of each component in the finished homogeneous thermoset copolymers. After passage through this zone, the mixture resulting therefrom may be continuously withdrawn and utilized to coat and/or impregnate a substrate or reinforcement. The coated or impregnated substrate or reinforcement may thereafter be continuously charged to a curing zone where it is subjected to a partial cure by passage through this zone which is maintained at varying operating temperatures for a predetermined period of time. After passage through the zone, the resulting prepreg material is continuously withdrawn and passed to storage. The prepreg can then be layed up as sheets with or without a metal such as copper foil as an electrical or thermal conductor, and pressed in a predetermined number of sheets to form the desired laminate or circuit board matrix.

It is also within the scope of this invention that the circuit board precursor may be prepared by a solventless continuation lamination process. When this type of process is employed, the solid resin blend comprising at least one poly(vinyl benzyl ether) of a polyphenol and a dicyanate ester of a polyether of a polyphenol or a polyaromatic cyanate compound is used to impregnate a reinforcement such as glass cloth which is continuously fed through an appropriate apparatus. The reinforcement such as the glass cloth may pass through this apparatus in a single ply or, if so desired, in a predetermined number of plies, one criteria being that each ply is impregnated with the resin blend. As an alternative, it is also contemplated that, if the laminate is to be used as a circuit board, one or both sides of the laminate may be covered with a metallic coating such as copper foil. The laminate is then passed through the apparatus under predetermined conditions of temperature and pressure so as to provide a finished and cured laminate which emerges from the apparatus. This metal-covered laminate or uncovered laminate may then be cut into desired sizes and utilized, as hereinbefore set forth, as a circuit board in various electric or electronic devices.

Regardless of the method which is utilized to form the desired copolymer, it is contemplated that the two components of the final composition of matter may be present in various weight ratios. In the preferred embodiment of the invention, the poly(vinyl benzyl ether) of a polyphenol will be present in the finished composite in an amount in the range of from about 10% to about 90% by weight of the finished composite, while conversely the dicyanate ester of a polyether of a polyphenol or a polyaromatic cyanate compound will be present

in an amount in the range of from about 90% to about 10% by weight of the composite. While these weight ratios are preferred, it is also contemplated within the scope of this invention that either of the two components may be present in the finished composite in either a greater amount or a lesser amount, depending upon the particular properties which are desired to be possessed by the finished composite.

In addition to the aforementioned favorable characteristics which are possessed by the homogeneous thermoset copolymers of the present invention, another advantage in utilizing these copolymers as components of a laminate is when employing a halogenated derivative of a poly(vinyl benzyl ether) of a polyphenol or a dicyanate ester of a polyether of a polyphenol or a polyaromatic cyanate compound as a component of the copolymer. The presence of these halogenated derivatives, and especially the brominated or chlorinated derivative, will introduce a desired property enhancement to a substrate or reinforcement in that the laminate may then meet certain flammability requirements such as UL 94 flammability tests.

The following examples are given for purposes of illustrating the novel homogeneous thermoset copolymers of the present invention which possess the aforementioned desirable properties and to a method for the preparation thereof. However, it is to be understood that these examples are given merely for purposes of illustration and that the present invention is not necessarily limited thereto.

EXAMPLE I:

To form a desired homogeneous thermoset copolymer, 21.9 grams of styrene capped bisphenol A [i.e. a di(vinyl benzylether) of bisphenol A] and 21.9 grams of the dicyanate ester of bisphenol A were dissolved at room temperature in 26 grams of dimethyl formamide. A stable homogeneous varnish resulted in which no recrystallization occurred after allowing the resin to stand for a period of 24 hours at ambient temperature. The resin was coated on a glass cloth substrate at room temperature. The solvent was removed by treatment in an oven at a temperature of 171°C for 7 minutes. The resulting prepreg was pressed at a temperature of 177°C and a pressure of 200 pounds per square inch gauge (1378 kPa gauge) for a period of 1 hour to form a laminate. The laminate was then subjected to a post-cure at a temperature of 230°C for a period of 3 hours. Analysis by DSC of the post-cured laminate showed a T_g of greater than 240°C.

EXAMPLE II:

In this example, 27.5 grams of styrene-capped bisphenol A and 27.4 grams of a chain-advanced dicyanate ester of bisphenol A, sold under the trade name RDX 76680 by the Celanese Corporation, were dissolved in 50 grams of dimethyl formamide at room temperature. As in the above example, no recrystallization of the resulting resin occurred after allowing the resin to stand at ambient temperature for a period of 24 hours. The resin was then used to impregnate a glass cloth after which the dimethyl formamide solvent was removed by treatment of the impregnated cloth in an oven at a temperature of 171°C for a period of 7 minutes. The prepreg was then pressed at a temperature of 177°C and a pressure of 200 pounds per square inch gauge (1378 kPa gauge) for a period of 1 hour to form a laminate. This laminate was then post-cured at a temperature of 230°C for a period of three hours. Analysis of the post-cured laminate by DSC showed a T_g of greater than 240°C.

EXAMPLE III:

In a manner similar to that set forth in the above examples, equal amounts of styrene-capped bisphenol A and the dicyanate ester of tetrabromo-substituted-bisphenol A are dissolved in a suitable solvent such as dimethyl formamide at a temperature of about 20°C to form a homogeneous thermoset copolymer. This copolymer is then used to impregnate a substrate such as a glass cloth which is thereafter pressed at an elevated temperature to form a laminate which is then post-cured also at an elevated temperature to form the desired product.

EXAMPLE IV:

A series of homogeneous thermoset copolymers were prepared by dissolving 25 grams of styrene-capped tetrabromo bisphenol A [i.e. a di(vinyl benzylether) of tetrabromo bisphenol A] in 15 grams of methyl ethyl ketone with heating. A polyaromatic cyanate compound in which the bridge moiety comprises dicyclopentadiene sold under the trade name "Dow XU-71787" by the Dow Chemical Company was also dissolved in methyl ethyl ketone in varying amounts so that the weight ratio of the polyaromatic cyanate compound to the styrene terminated tetrabromo bisphenol A was in a weight ratio of 64:36, 50:50 and 40:60, respectively. The two solutions were blended following which 4 grams

of dicumyl peroxide dissolved in 5 grams of methyl ethyl ketone were added to the blend. To this blend, 0.02 gram of zinc octoate solution was added. The resulting copolymer in the form of a resin was used to saturate a glass cloth substrate at room temperature. The solvents were removed by treating the substrate in an oven at a temperature in the range of from 100° to 180°C to drive out the solvent and B-stage the resin. The B-staged prepreg was then stacked between copper foil and pressed at a temperature of 150°C for a period of one hour followed by one hour at 177°C to form a laminate. The pressed laminate was then subjected to a post-cure at a temperature of 204°C for a period of 4 hours. The properties of the three laminates are set forth in the table below in which Laminate A had a weight ratio of polyaromatic cyanate compound to styrene terminated tetrabromo Bisphenol A of 64:36; Laminate B had a weight ratio of 50:50 and Laminate C had a weight ratio of 40:60. Laminate D comprised a commercial epoxy laminate.

Dow 71787 - Styrene Terminated TetrabromoBPA Laminate Properties

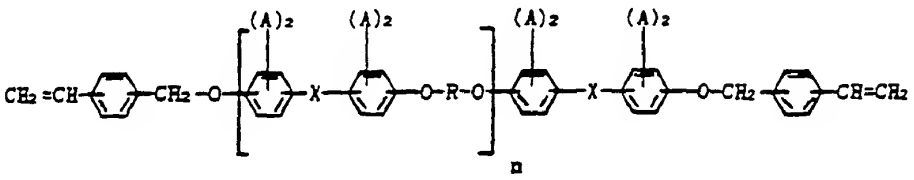
	A	B	C	D
Dielectric Constant (A)	3.3	3.8	3.7	4.2-4.3
Dissipation Factor (A)	0.008	0.01	0.01	0.01
T_g (°C)	248	272	304	125
Water Absorption (D-24/100 % gain)	0.70	n/a	n/a	1-1.2
Methylene Chloride Absorption (% gain)	1.5	n/a	0.5	1-1.5
UL-94 Flammability Rating (A)	V-1	V-0	V-0	V-0
Resin Content (%)	66	38.2	51	50

It is noted from a comparison of results in the above table that the dielectric constant of each of the laminates of the present invention was less than the dielectric constant of the commercial laminate and, in addition, the T_g of these laminates was considerably higher than that of the commercial laminate. This combination of low dielectric constant and high glass transition temperature

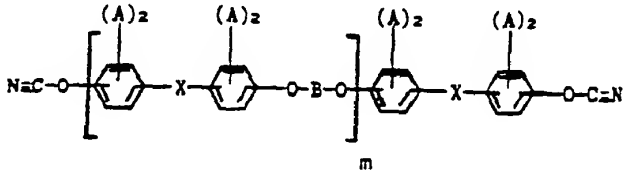
clearly demonstrates the superior qualities of a laminate of the present invention when compared to commercial products now in use.

WE CLAIM AS OUR INVENTION:

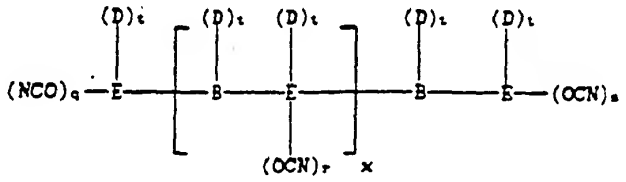
1. A homogeneous thermoset copolymer of a poly(vinyl benzyl ether) of a polyphenol having the structure:



in which X is selected from the group consisting of CH_3-$\overset{|}{\underset{|}{C}}$-$CH_3$, SO_2, O, S, and CH_2 radicals, R is selected from the group consisting of -CH_2-C_6H_4-CH_2-, $(CH_2)_b$ in which b ranges from 1 to about 6, -CH_2-CH=CH-CH_2- and -CH_2-C≡C-CH_2- radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and n has an average value in the range of from about 0 to about 20 and a cyanate compound selected from the group consisting of a dicyanate ester of a polyether of a polyphenol having the general formula:



in which X is selected from the group consisting of CH_3-$\overset{|}{\underset{|}{C}}$-$CH_3$, SO_2, O, S, and CH_2 radicals, B is selected from the group consisting of cyclic and polycyclic aliphatic radicals containing from 5 to about 22 carbon atoms, -CH_2-C_6H_4-CH_2-, $(CH_2)_b$ in which b ranges from 1 to about 6, -CH_2-CH=CH-CH_2- and -CH_2-C≡C-CH_2- radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and m has an average value in the range of from about 0 to about 20 and a polyaromatic cyanate compound having the general formula:



in which B is a polycyclic aliphatic radical containing from about 7 to about 20 carbon atoms, D is any nonactive hydrogen-containing substituent, E is an aromatic radical, q, r and s are independently in each occurrence the integers 0, 1, 2, or 3, with the proviso that the sum of q, r and s is greater than or equal to 2, t is independently in each occurrence an integer of between about 0 and 4, and x is a number between about 0 and 5.

2. The homogeneous thermoset copolymer as set forth in Claim 1 in which said poly(vinyl benzyl ether) of a poly phenol is present in said copolymer in an amount in the range of from about 10% to about 90%.

3. The homogeneous thermoset copolymer as set forth in Claim 1 in which said dicyanate ester of a polyether of a polyphenol or polyaromatic cyanate compound is present in said copolymer in an amount in the range of from about 90% to about 10%.

4. The homogeneous thermoset copolymer as set forth in Claim 1 in which A is hydrogen.

5. The homogeneous thermoset copolymer as set forth in Claim 1 in which A is bromine.

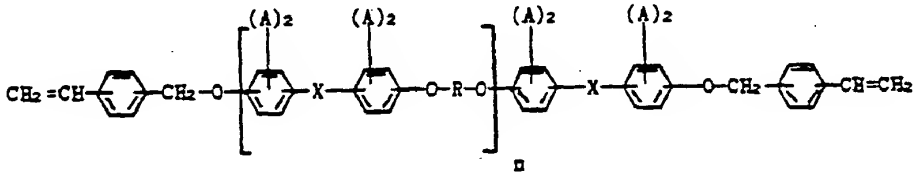
6. The homogeneous thermoset copolymer as set forth in Claim 1 in which A is methyl.

7. The homogeneous thermoset copolymer as set forth in Claim 1 in which D is hydrogen.

8. The homogeneous thermoset copolymer as set forth in Claim 1 in which D is bromine.

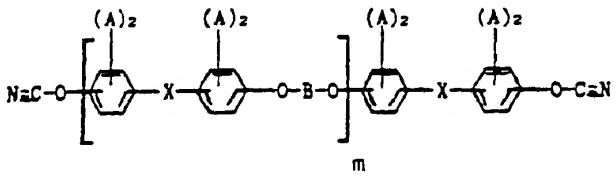
9. The homogeneous thermoset copolymer as set forth in Claim 1 in which B is a dicyclopentadiene radical.

10. A process for the production of a homogeneous thermoset copolymer which comprises reacting a poly(vinyl benzyl ether) of a polyphenol having the structure:

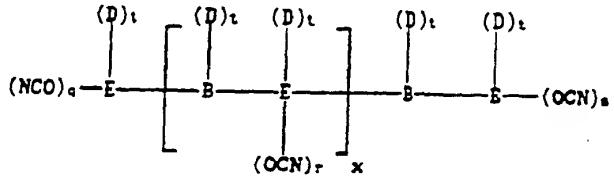


in which X is selected from the group consisting of CH_3-$\overset{|}{C}$-CH_3, SO_2, O, S, and CH_2 radicals, R is selected from the group consisting of $-CH_2-C_6H_4-CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2-CH=CH-CH_2-$ and $-CH_2-C\equiv C-CH_2-$ radicals, A is independently selected from the group consisting

of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and n has an average value in the range of from about 0 to about 20 with a cyanate compound selected from the group consisting of a dicyanate ester of a polyether of a polyphenol having the structure:



in which X is selected from the group consisting of CH_3-$\overset{|}{\underset{|}{C}}$-$CH_3$, SO_2, O, S, and CH_2 radicals, B is selected from the group consisting of cyclic and polycyclic aliphatic radicals containing from 5 to about 22 carbon atoms, $-CH_2-C_6H_4-CH_2-$, $(CH_2)_b$ in which b ranges from 1 to about 6, $-CH_2-CH=CH-CH_2-$ and $-CH_2-C\equiv C-CH_2-$ radicals, A is independently selected from the group consisting of hydrogen, chlorine, bromine, fluorine, alkyl, alkoxy and phenyl radicals and m has an average value in the range of from about 0 to about 20 and a polyaromatic cyanate compound having the general formula:



in which B is a polycyclic aliphatic radical containing from about 7 to about 20 carbon atoms, D is any nonactive hydrogen-containing substituent, E is an aromatic radical, q, r and s are independently in each occurrence the integers 0, 1, 2, or 3, with the proviso that the sum of q, r and s is greater than or equal to 2, t is independently in each occurrence an integer of between about 0 and 4, and x is a number between about 0 and 5 at reaction conditions, and recovering the resultant homogeneous thermoset copolymer.

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 87/03368

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC[4]: C 08 G 73/00; C 08 G 85/00

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
IPC[4]	C 08 G

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
A	US, A, 4116946 (H.D. JAKOB) 26 September 1978 ---	

* Special categories of cited documents: [10]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
24th May 1988	22 JUN 1988
International Searching Authority	**Signature of Authorized Officer**
EUROPEAN PATENT OFFICE	P.C.G. VAN DER PUTTEN

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO. US 8703368 SA 20869

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
The members are as contained in the European Patent Office EDP file on 09/06/88
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A- 4116946	26-09-78	NL-A- 7706914	28-12-77
		BE-A- 856012	23-12-77
		DE-A,C 2628417	05-01-78
		FR-A,B 2361444	10-03-78
		GB-A- 1544162	11-04-79
		JP-A- 53000300	05-01-78
		AT-B- 358287	25-08-80

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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82